Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

High efficiency and long lifetime orange-red emitting thermally activated delayed fluorescent organic light emitting diodes by donor and acceptor engineering

Shantaram Kothavale, Won Jae Chung, Jun Yeob Lee*

School of Chemical Engineering, Sungkyunkwan University

2066, Seobu-ro, Jangan-gu, Suwon, Gyeonggi, 440-746, Korea

E-mail: leej17@skku.edu

Table of contents

Experimental

General information

1-(4-bromophenyl)-2-phenylethane-1,2-dione, 4,5-diaminophthalonitrile, tris-tert-butylphosphine ($P(t\text{-butyl})_3$), and palladium acetate ($Pd(OAc)_2$) were purchased from P&H tech. p-toluenesulphonic acid (PTSA) and copper cyanide (CuCN) were purchased from Alfa Aesar Co. Caesium carbonate (Cs_2CO_3), anhydrous ethanol, *N,N*-Dimethylformamide (DMF), anhydrous toluene, and acetic acid were obtained from Duksan Sci. Co. All these chemicals were used without further purification. Column chromatography (Silica Gel 60, 230−400 mesh, Merck) purified both the TADF emitters were further purified by sublimation (10^{-3} Torr at 300 °C) before applying for OLED devices. The ultraviolet-visible (UV-vis) absorption spectra and photoluminescence (PL) spectra were recorded using UV-vis spectrophotometer (JASCO, V-730) and fluorescence spectrophotometer (PerkinElmer, LS-55) respectively. CV measurement was carried out using Ivium Tech., Iviumstat instrument in dichloromethane solution with scan rate at 100 mV/s. The glassy carbon, platinum wire and Ag/AgCl were used as working, counter and reference electrode respectively. Internal standard was ferrocenium/ferrocene couple and supporting electrolyte was 0.1 M tetrabutylammonium perchlorate ($TBAClO_4$). Absolute photoluminescence quantum yields (PLQYs) of 1 wt % doped polystyrene film were measured with a Hamamatsu Quantaurus-QY C11347-11 spectrometer and the transient photoluminescence decay characteristics of solid film samples were recorded using a Quantaurus-Tau fluorescence lifetime measurement system (C11367-31, Hamamatsu Photonics). The ^{1}H and ^{13}C nuclear magnetic resonance (NMR) spectra were recorded on a Avance-500 (Bruker, 500 MHz) spectrometer using deuterated chloroform ($CDCl_3$) solvent. Chemical shifts of the ^{1}H and ^{13}C NMR signals were quoted relative to tetramethylsilane ($\delta = 0.00$). All coupling constants are reported in Hertz. The mass spectra were recorded using a Advion, Expresion LCMS spectrometer in APCI mode. TD-DFT calculations was carried out using the Gaussian 09 package and Becke's three parameter exchange functional B3LYP with basis set of 6-31G (d).

Table S1. Electroluminescence performance of the lifetime based TADF OLED devices at 1 % doping concentration.

Emitter	λ_{EL} [nm]	CE_{max} [cd A^{-1}]	PE_{max} [lm W^{-1}]	EQE_{max} [%]	CIE (x , y)
6,7-DCNQx-DICz	571	56.8	44.2	19.9	(0.47, 0.51)
5,8-DCNQx-DICz	597	22.8	17.3	10.6	(0.55, 0.45)

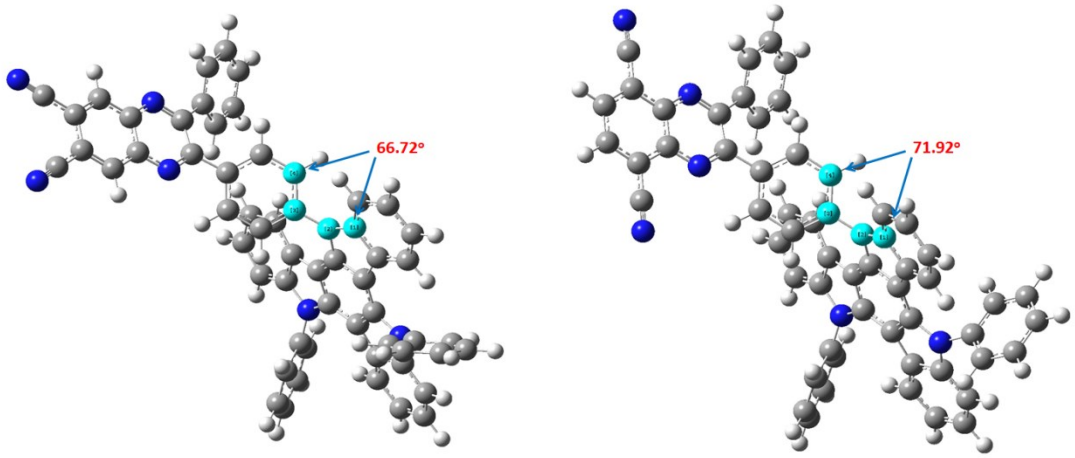

Figure. S1. DFT optimized structures of **6,7-DCNQx-DICz** and **5,8-DCNQx-DICz** with their dihedral angles between donating unit and phenyl ring spacer at ground state.

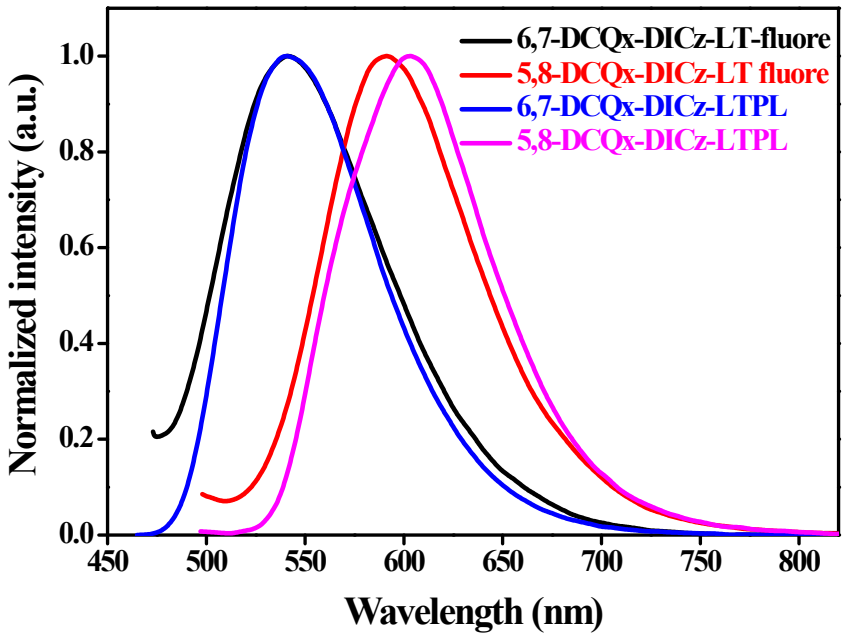

Figure. S2. Fluorescence and phosphorescence spectra of **6,7-DCNQx-DICz** and **5,8-DCNQx-DICz** at 77 K in toluene solvent.

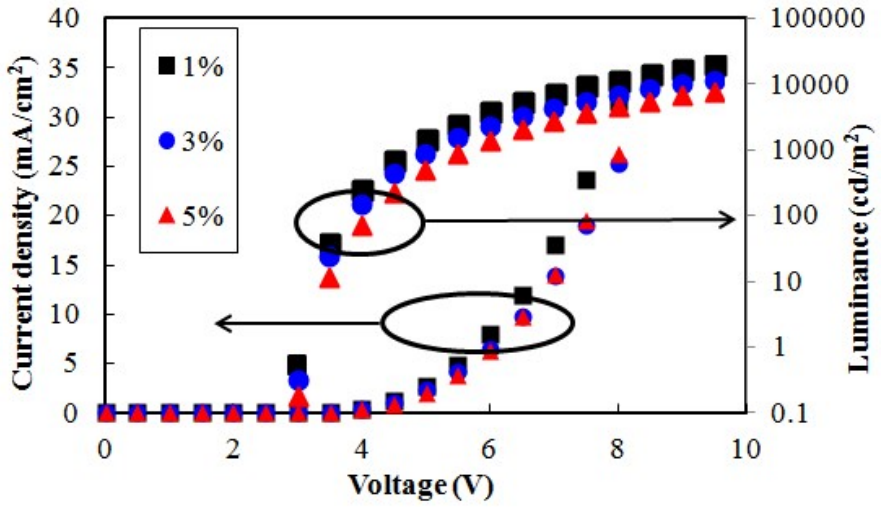

Figure. S3. Current density−voltage−luminance (J−V−L) curves of **6,7-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

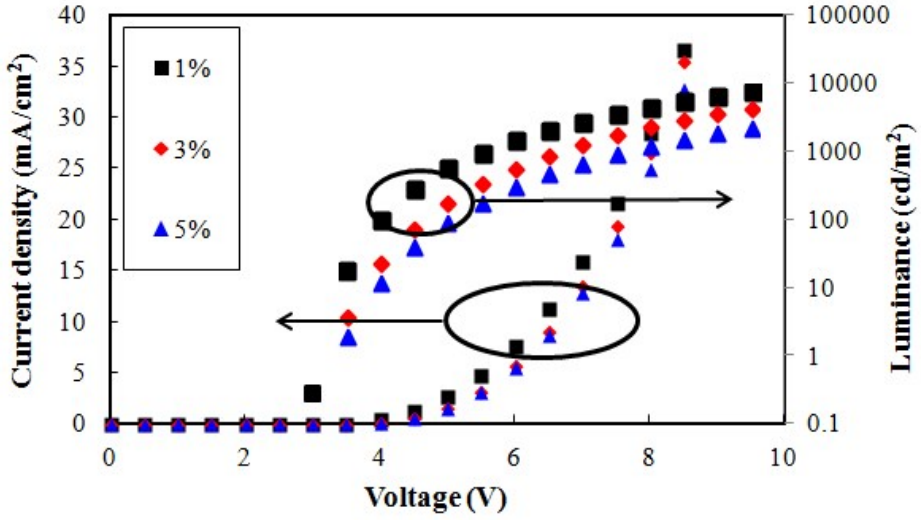

Figure S4. Current density−voltage−luminance (J−V−L) curves of **5,8-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

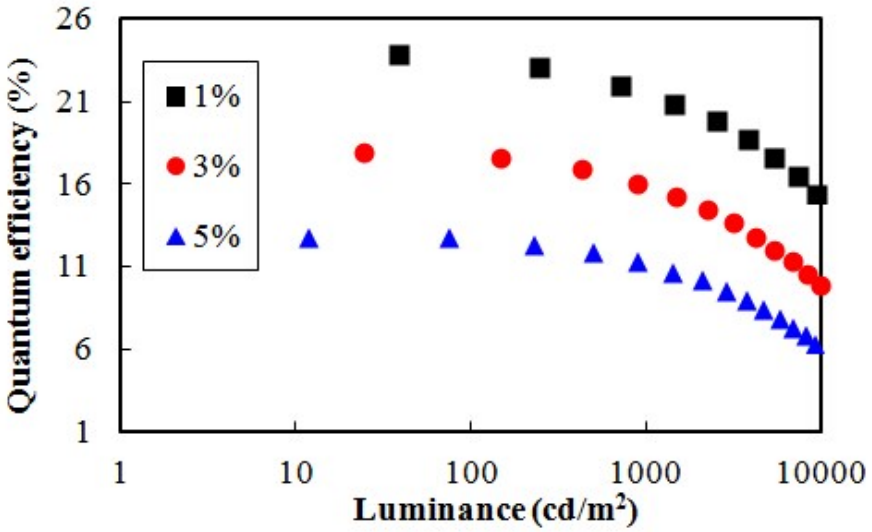

Figure S5. EQE−luminance curves of **6,7-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

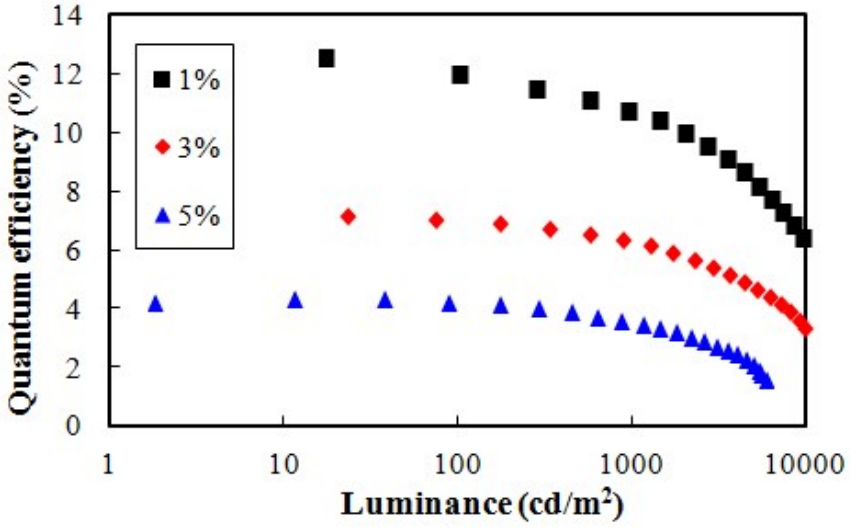

Figure S6. EQE−luminance curves of **5,8-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

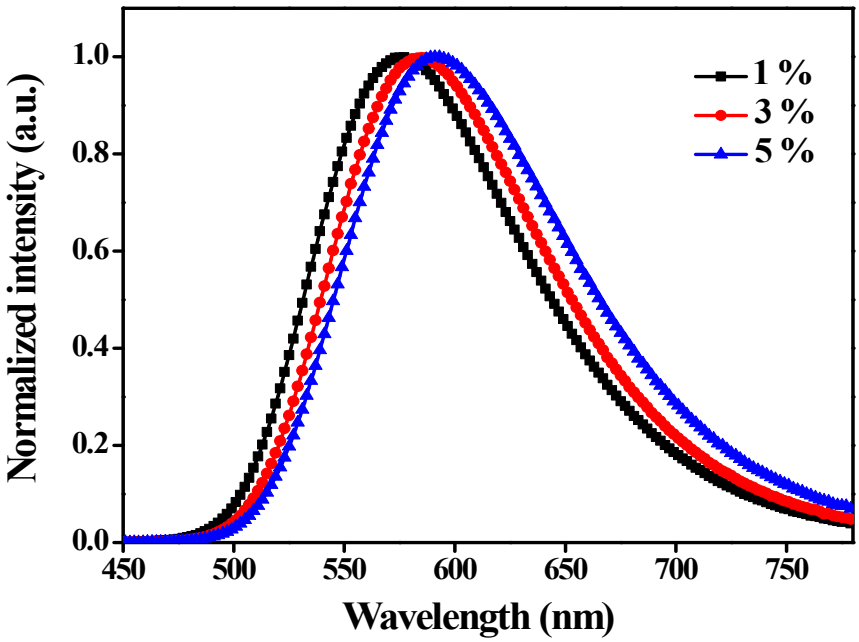

Figure S7. EL spectra of **6,7-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

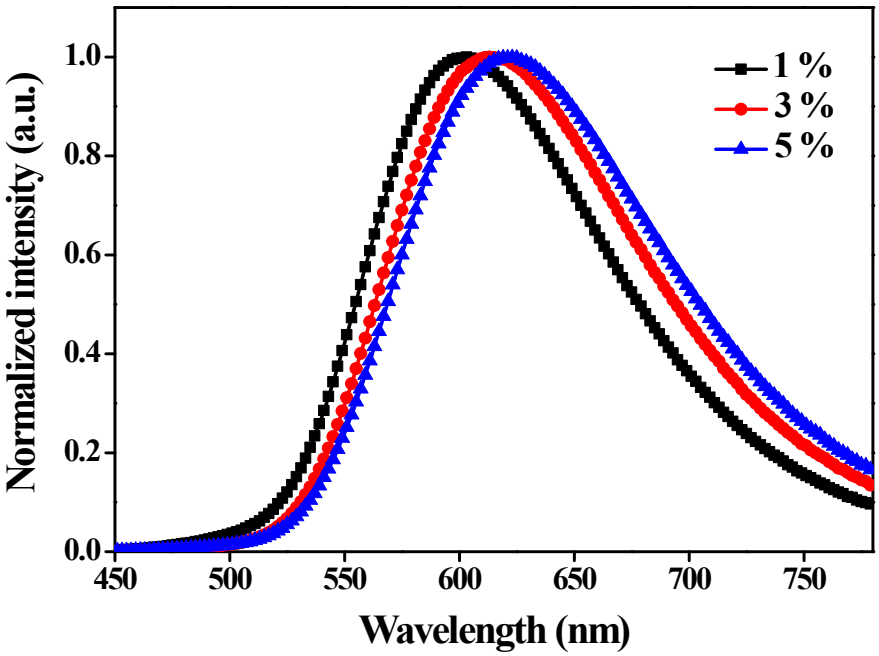

Figure S8. EL spectra of **5,8-DCNQx-DICz** at 1, 3 and 5 % doping concentration.

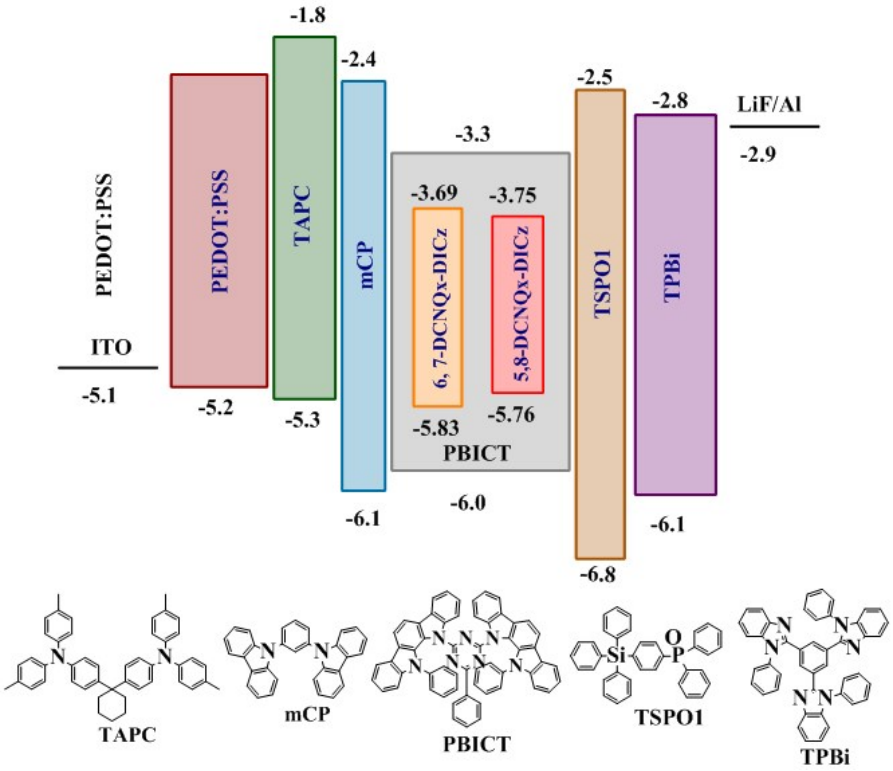

Figure S9. Efficiency based device diagram with chemical structures of the material used.

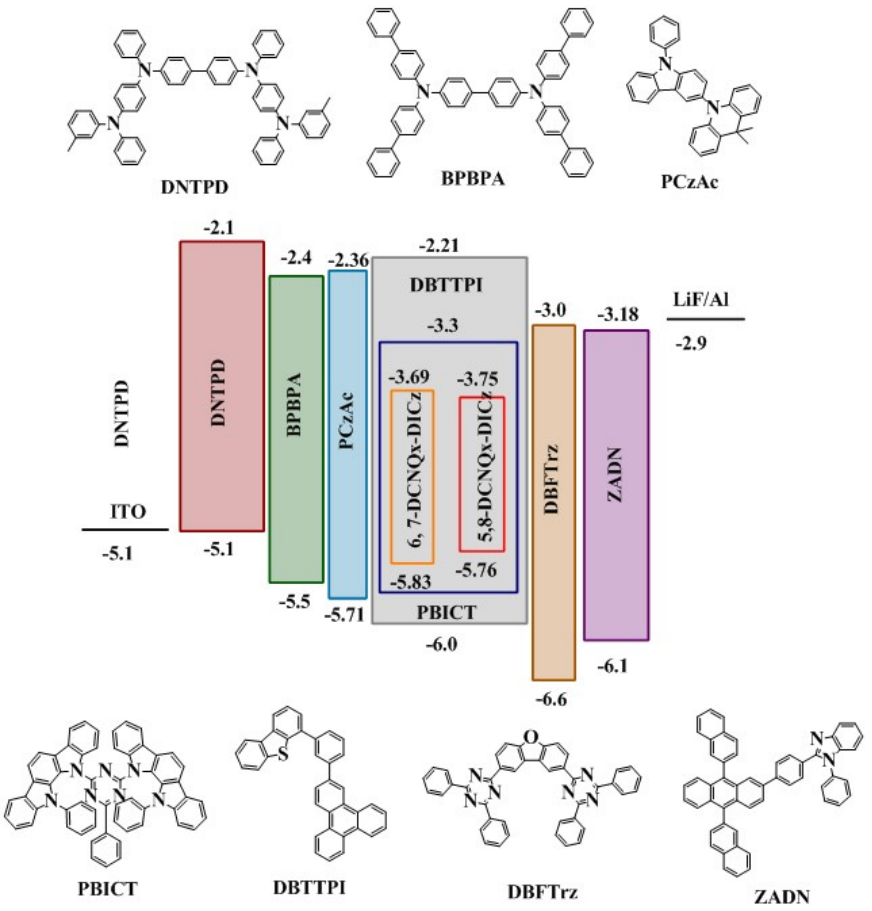

Figure S10. Lifetime based device diagram with chemical structures of the material used.

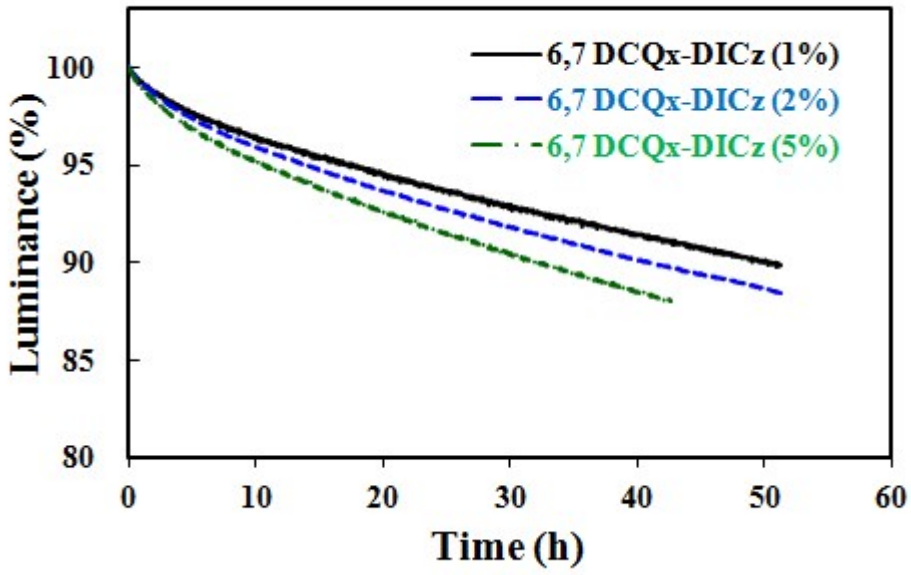

Figure S11. Lifetime curves of **6,7 DCQx-DICz** at 1, 3, and 5 % doping concentration.

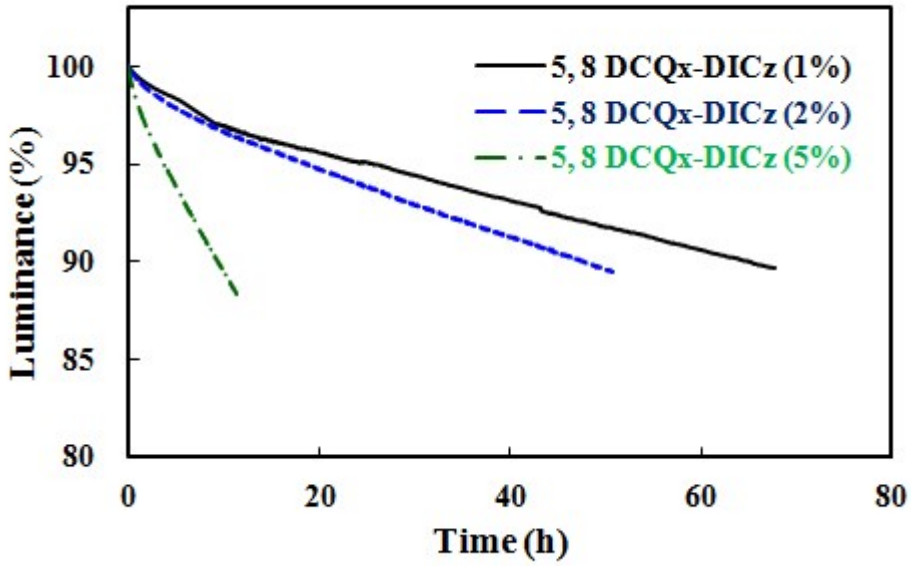

Figure S12. Lifetime curves of **5,8 DCQx-DICz** at 1, 3, and 5 % doping concentration.

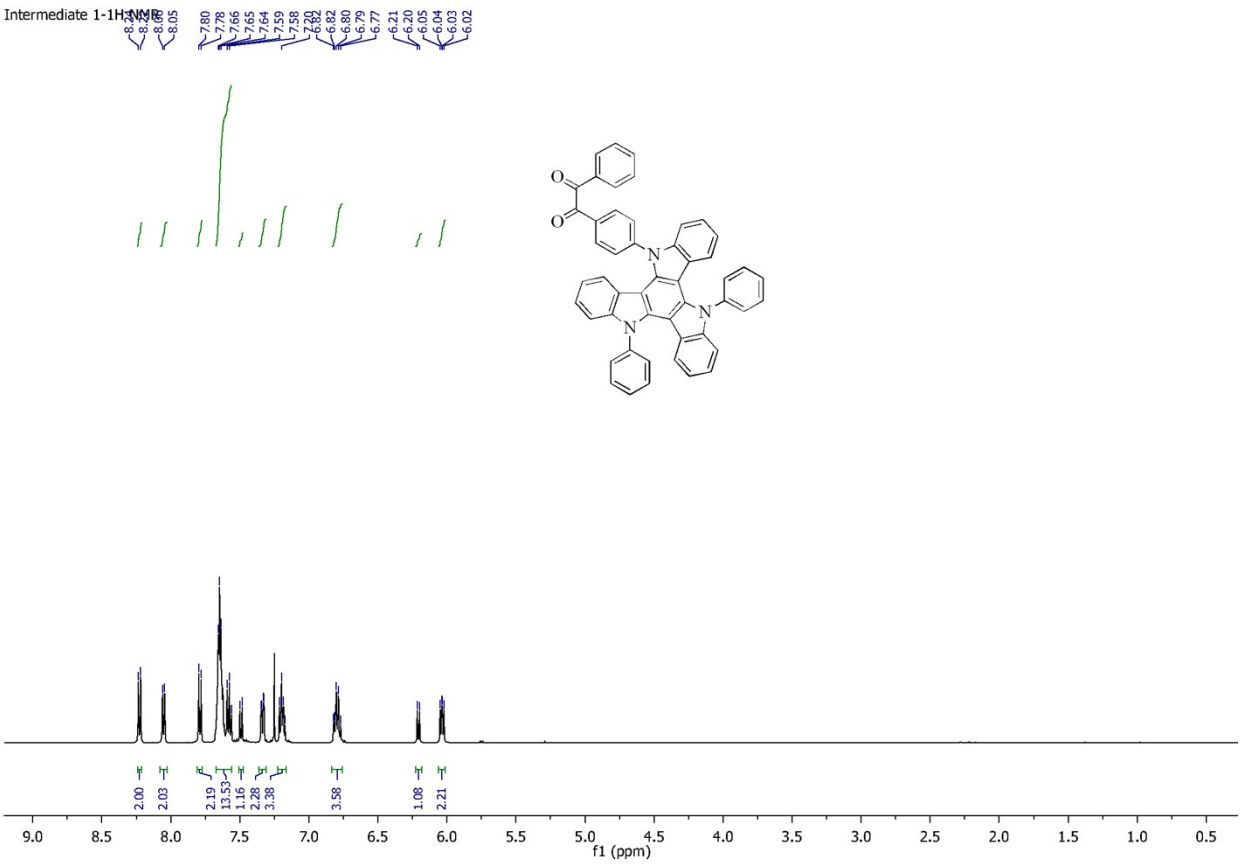

Figure S13. ^{1}H NMR spectrum of intermediate 1.

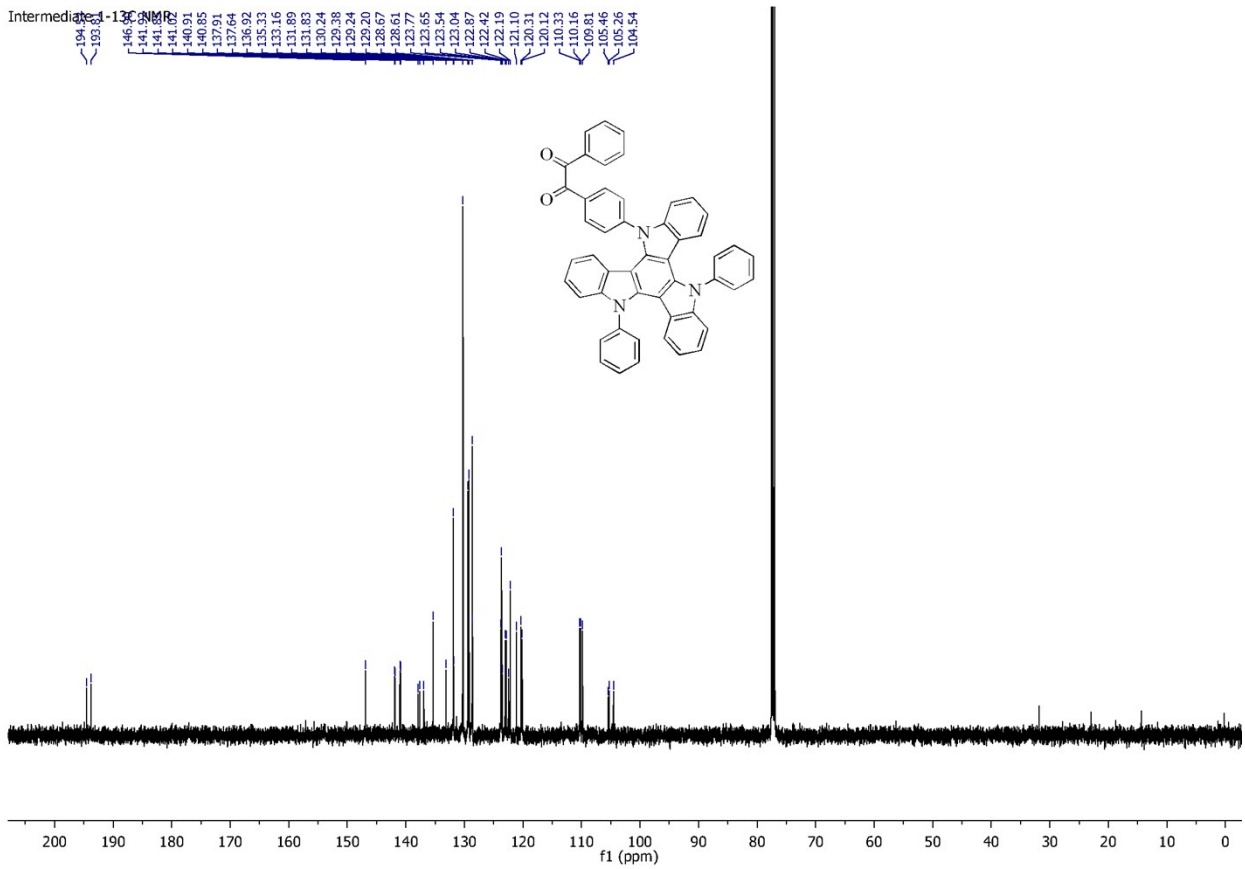

Figure S14. ^{13}C NMR spectrum of intermediate 1.

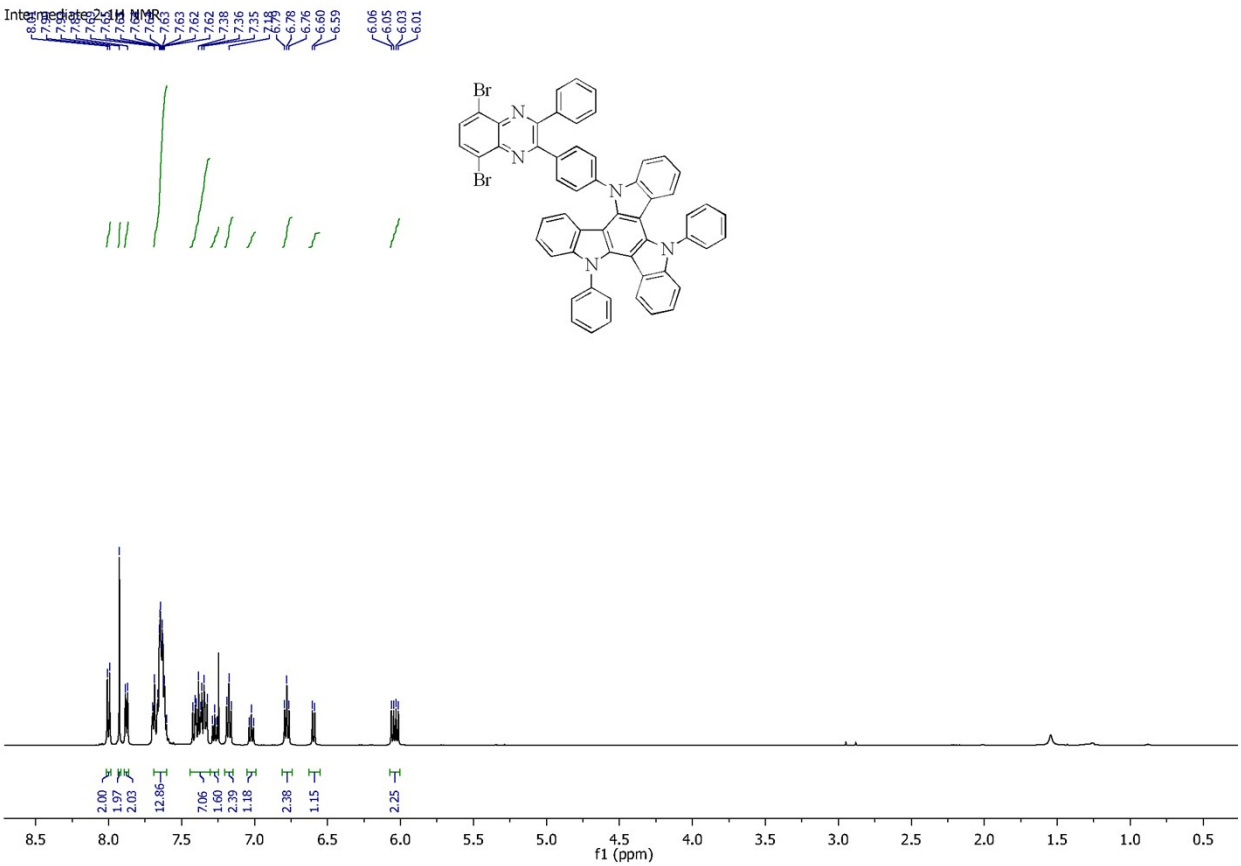

Figure S15. ^{1}H NMR spectrum of intermediate 2.

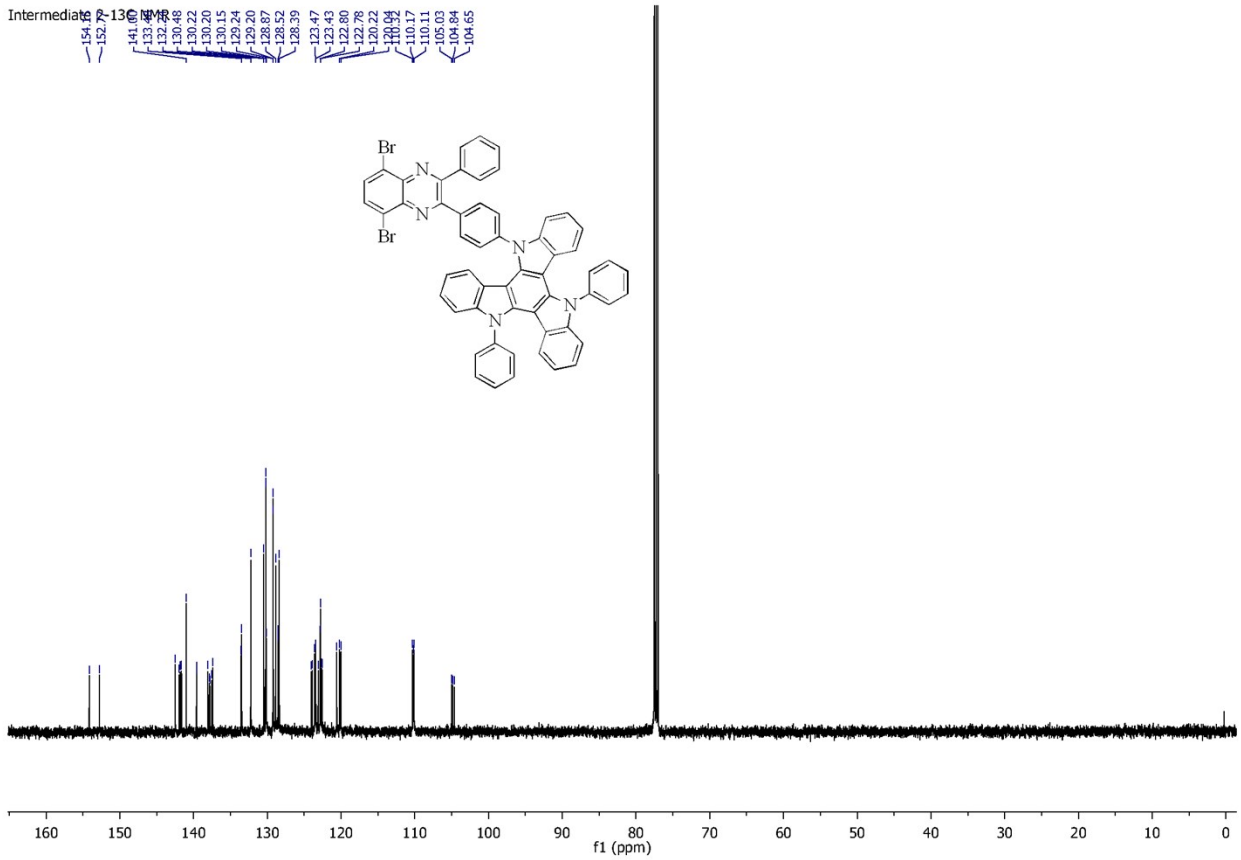

Figure S16. ^{13}C NMR spectrum of intermediate 2.

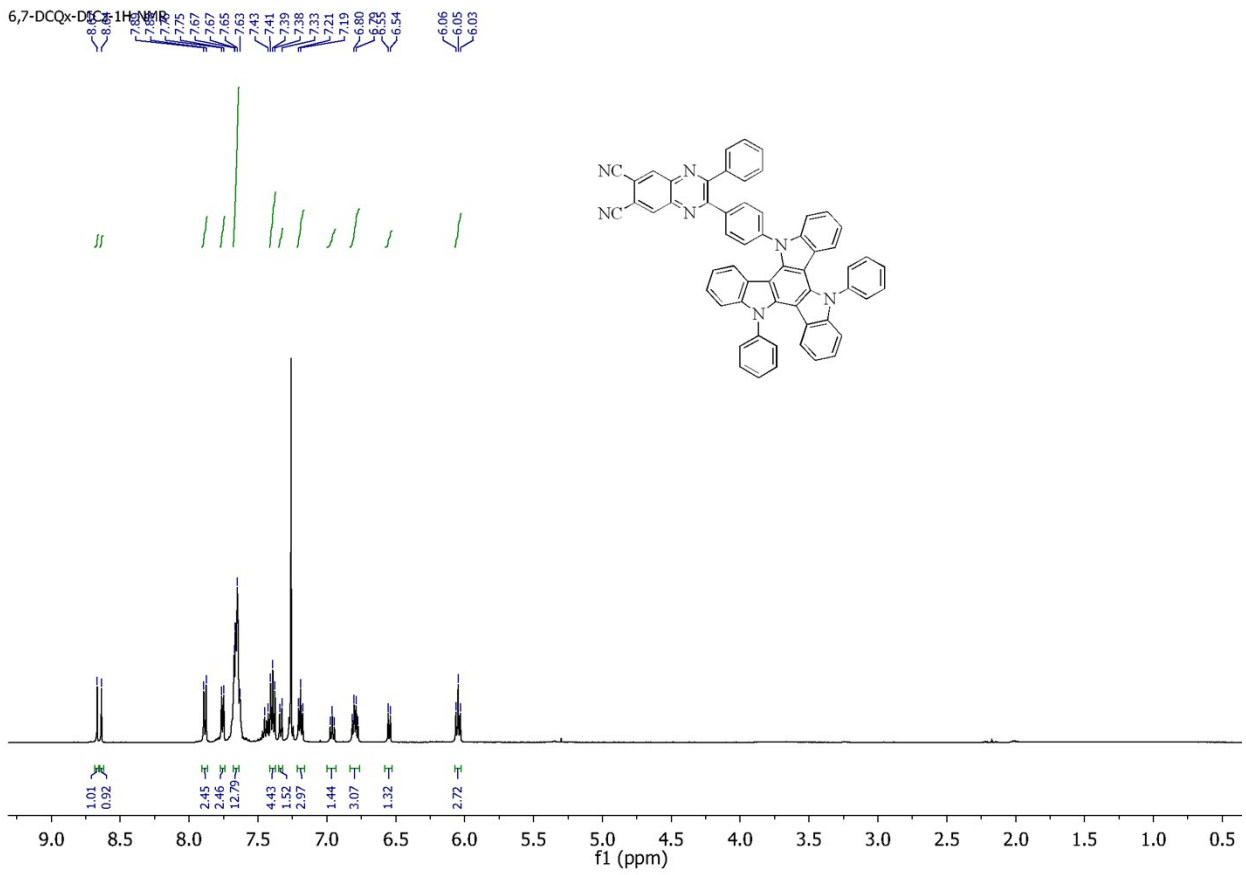

Figure S17. ^{1}H NMR spectrum of **6,7-DCNQx-DICz.**

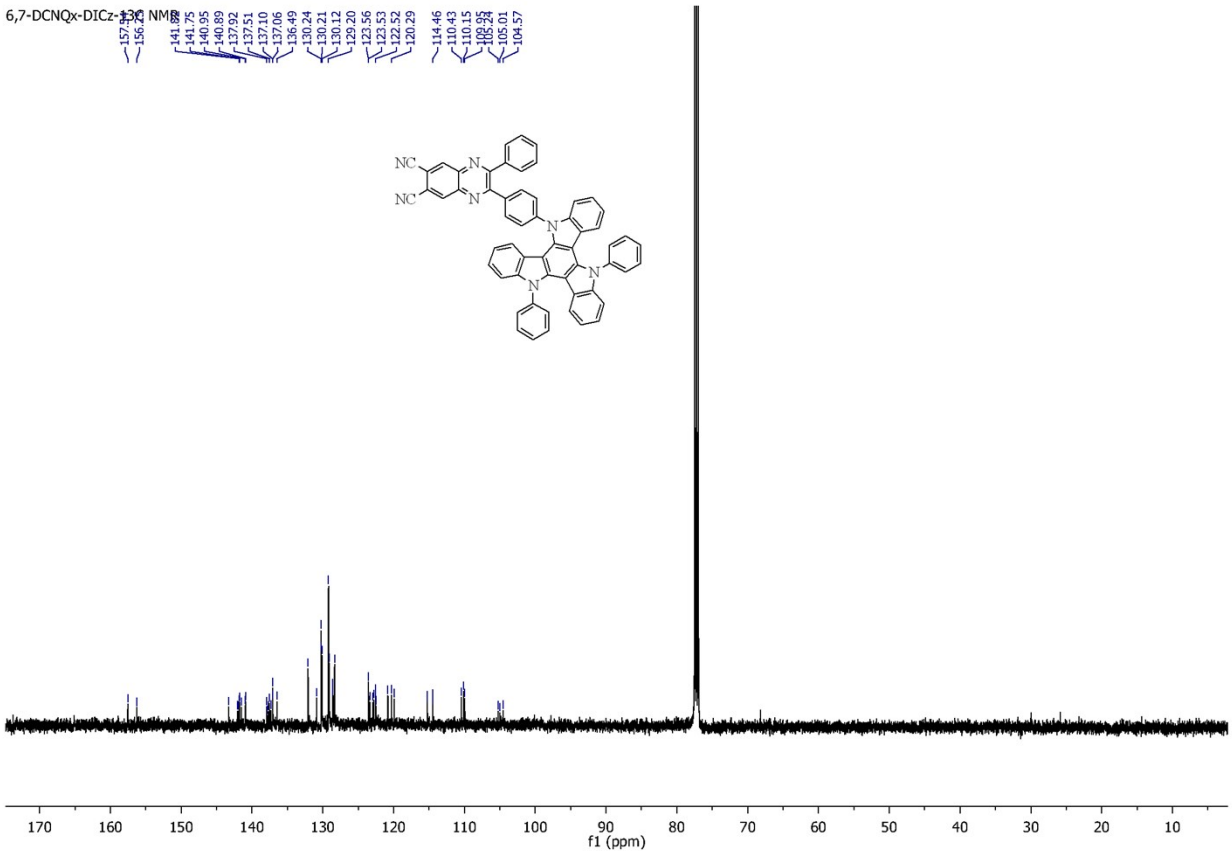

Figure S18. ^{13}C NMR spectrum of **6,7-DCNQx-DICz.**

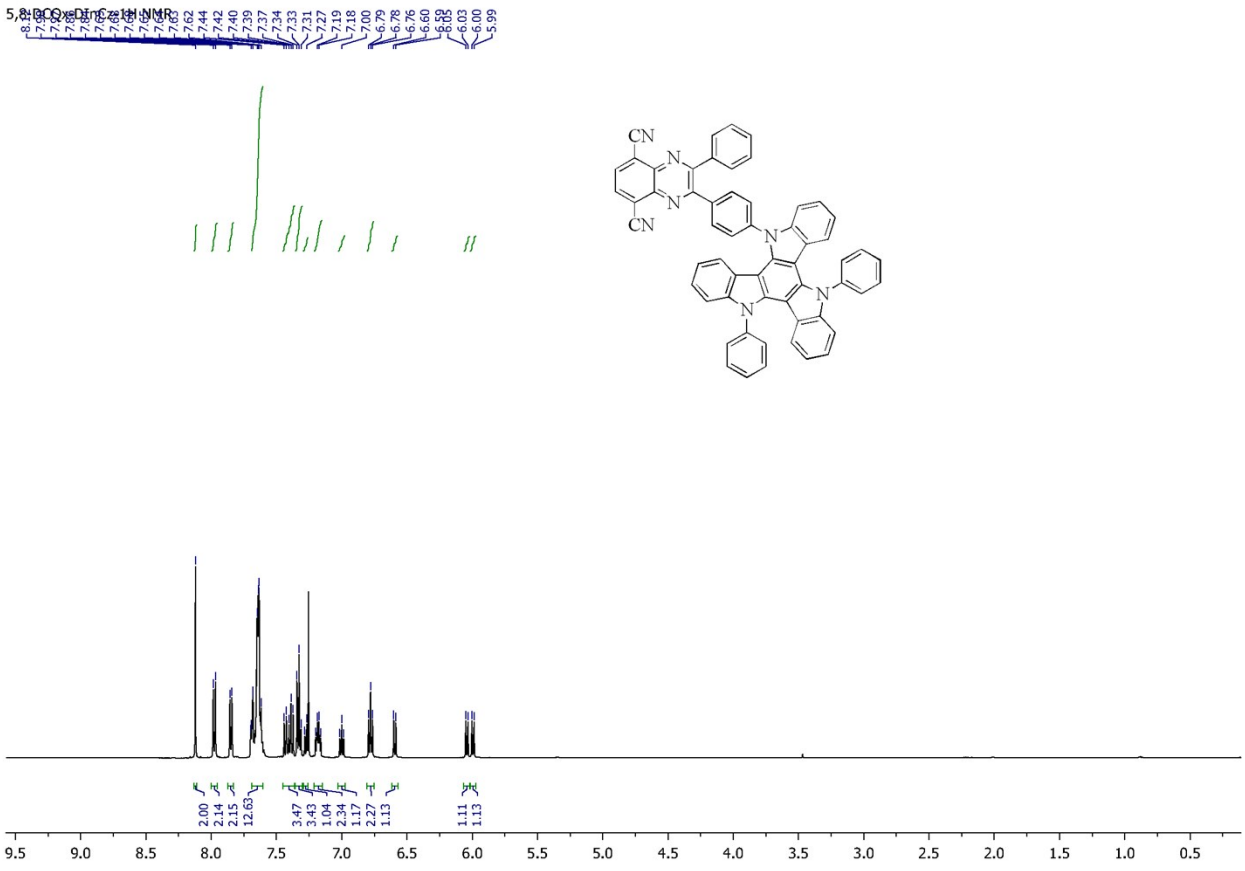

Figure S19. ^{1}H NMR spectrum of **5,8-DCNQx-DICz.**

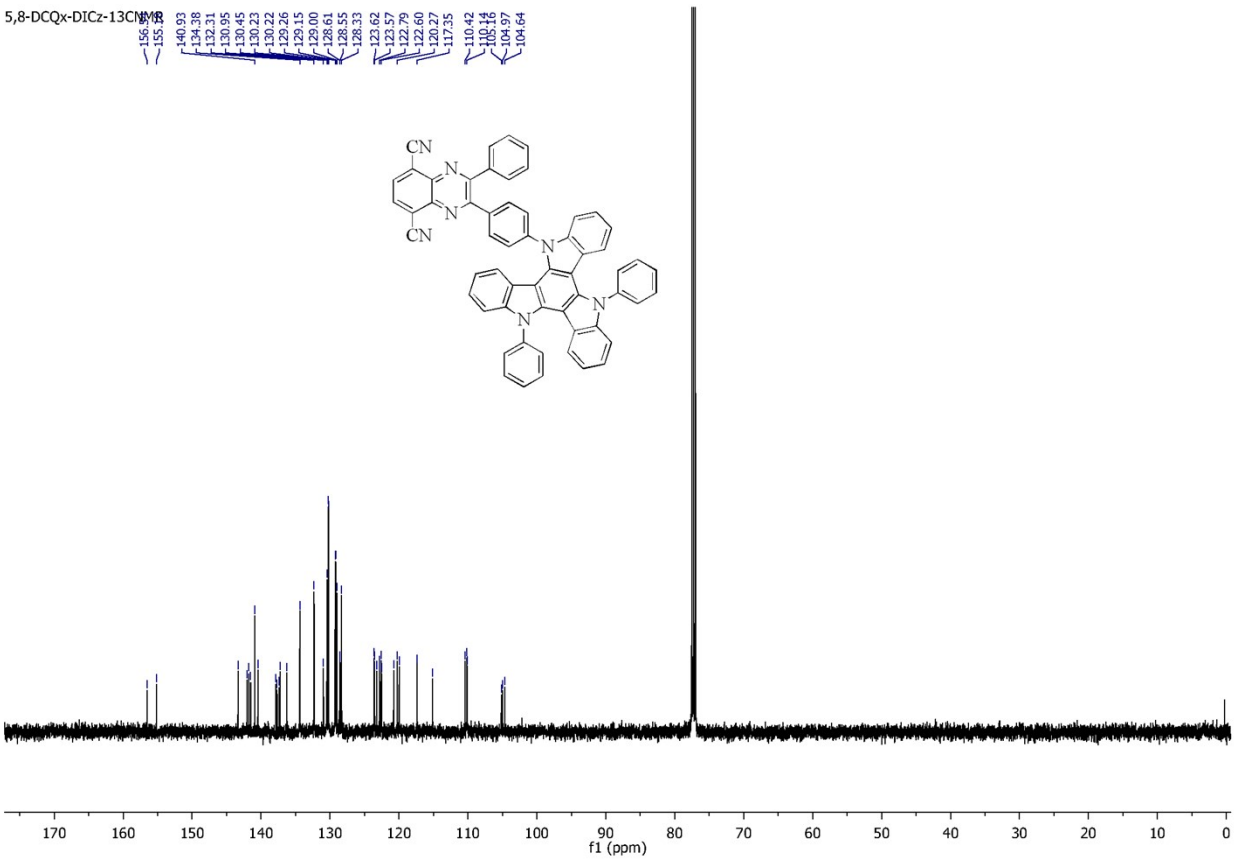

Figure S20.[13]C NMR spectrum of **5,8-DCNQx-DICz.**